A new universality class of aging in glassy systems

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Glassy systems are ubiquitous in nature, and are characterized by slow relaxations to equilibrium without a typical timescale, aging and memory effects. Understanding these is a long-standing problem in physics. We study the aging of the electron glass, a system showing remarkable slow relaxations in the conductance. We find that the broad distribution of relaxation rates leads to a universal relaxation of the form $\log(1 + t_w/t)$ for the usual aging protocol, where t_w is the length of time the perturbation driving the system out-of-equilibrium was on, and t the time of measurement. These results agree well with several experiments performed on different glasses, and examining different physical observables, for times ranging from seconds to several hours. The suggested theoretical framework appears to offer a paradigm for aging in a broad class of glassy materials.

Aging is one of the most distinct characteristics of glasses. Crudely speaking, it is the dependence of the form of the relaxation to equilibrium on the time an external perturbation was acting on the system. Aging and slow relaxations have been experimentally observed in various systems. These range from spin-glass [1], to structural glasses [2, 3], gels [4], vortices in superconductors [5], ferroelectrics [6], colloids and granular systems [7] and the electron-glass system [8]. Analytic models and simulations showing aging have been of considerate interest in the past decades [9]. In general, the return to equilibrium can be characterized by a function $F(t, t_w)$, where t_w, the 'waiting-time', is the time the perturbation was on (during which the system is being 'aged'), and t is the time from switching off the perturbation until the measurement. We refer to the complete protocol as an 'aging experiment'. Full (or simple) aging is said to hold when $F(t, t_w) = f(t_w/t)$.

Experiments performed on the electron glass system, measuring the conductance as a function of time when the system is perturbed, show remarkable data collapse when time is measured in units of t_w [8]. This proves experimentally that full aging is obeyed. For times $t \ll t_w$ a logarithmic relaxation is observed. At times t of order t_w deviations from the logarithm are seen. Similar results were obtained for granular aluminum [10] and structural glasses [3] which suggest that the physical mechanisms leading to such logarithmic relaxations may be ubiquitous. In this paper we suggest a model which should be applicable for several types of physical realizations, and yields the result that full aging indeed holds. Moreover, it gives under certain simplifying assumptions $f(x) \sim \log(1 + x)$, which fits the experimental data well for several decades in time.

The Model.- To start with, let us consider N localized disordered states and $M < N$ interacting electrons, with a coupling between the electrons and a phonon reservoir [11]. After ensemble averaging over the phonon reservoir, the averaged occupation numbers can take any value between 0 and 1, and their dynamics is described by a set of rate equations: $\frac{dn_i}{dt} = \sum_{j \neq i} \gamma_{j,i} - \gamma_{i,j}$, where due to the Coulomb interaction and the Pauli principle the rates $\gamma_{i,j}$, which describe the phonon-assisted transitions between electronic states, depend on the set of occupation numbers $\{n_i\}$. Linearization close to (one of the many) metastable states leads to an equation $\frac{d\vec{\delta n}}{dt} = A\vec{\delta n}$, where $\vec{\delta n}$ is a vector of the deviations from the metastable state, and A a matrix depending on the properties of the metastable state. Ref. [11] explains in detail the procedure, and shows that the distribution of eigenvalues of the matrix is given approximately by $P(\lambda) \sim 1/\lambda$ within a large frequency window, essentially arising from the exponential dependence of the matrix elements on the physical parameters (distance and energy difference). The $1/\lambda$ distribution leads to a logarithmic decay in time, as can be seen by Laplace transforming it.

From now on we consider quite generally a system whose dynamics is described by a linear relation $\frac{d\vec{\delta n}}{dt} = A \cdot \vec{\delta n}$, and with the eigenvalue distribution of the matrix A described approximately by $P(\lambda) \sim 1/\lambda$. The mean-field approximation leads to this result starting from a microscopic Hamiltonian, but we believe that the results should apply for a broader range of models. At the end of the paper we demonstrate this by applying the theory to the experimental data of Ref. [3].

We will assume that the system contains another external control parameter we shall call V_g, such as the gate voltage in the electron glass experiment [8]. Changing an external system parameter will shift the metastable states' positions. Thus, upon small changes in V_g, the linearized matrix A will slightly change. Let us consider the system in two metastable states a and b corresponding to values V_a and V_b respectively. We shall denote the linearized matrix A in the state $a(b)$ by A^a (A^b), and the eigenvectors $\vec{a}_q$ ($\vec{b}_q$). The metastable state a is

characterized by a configuration $\vec{n}_a$, and the state b by a configuration $\vec{n}_b = \vec{n}_a + \Delta \vec{n}$.

The experimental protocol is as follows:

I The gate voltage is V_a. The system is assumed to 'equilibrate' to a metastable state.

II The gate voltage is changed to $V_b = V_a + \delta V$, the system 'ages' for a time t_w: the system approaches the new equilibrium b.

III The gate voltage is changed back to a value V_a. The system is in the process of returning to the original equilibrium a.

The conductance is measured throughout the experiment. The measurement time is much shorter than the relevant relaxation times. For convenience we define $t = 0$ at the end of stage II, when the gate voltage is changed to its initial value V_a.

During stage I, the system reaches, presumably, one of the many metastable states (a), and lingers there. At the beginning of stage II ($t = -t_w$) we change the potential landscape by shifting suddenly the gate voltage from V_a to V_b. The occupations at this time are still equal to the metastable state occupations $\vec{n}_a$. *During* stage II it starts, however, to relax to the new metastable state b. The new metastable state b is not far (in phase space) from the original one a , but since the relaxation to it is composed of a broad range of timescales, the system takes a long time to fully equilibrate to state b. At the beginning of stage III ($t = 0$) the gate voltage is changed back to its initial value. During stage III, the system is relaxing back to a, the metastable state it was initially in [12]. This process is described schematically in Fig. 1, along with our theoretical predictions.

Let us proceed to the calculation. In stage I, the system is in the metastable configuration $\vec{n}_a$. At the beginning of stage II, the system is in the state $\vec{n}_a = \vec{n}_b - \Delta \vec{n}$. Working in the eigenbasis of b, $-\Delta \vec{n} = \sum_q c_q \vec{b}_q$. This equation determines the decomposition weights c_q (which are not simply the projections onto the eigenvectors, since the matrix A^b is not hermitian [11]). Therefore during stage II (the aging process), the configuration of the system is described by:

$$\vec{n}(t) = \vec{n}_b + \sum_q c_q \vec{b}_q e^{-\lambda_q^b(t+t_w)}, -t_w < t < 0, \quad (1)$$

where λ_q^b are the relaxation rates (the eigenvalues of the linearized matrices A^a and A^b are real and negative, corresponding to pure decay). Since the eigenvectors are localized [11], we can assume that c_q are random variables whose distributions and typical values do not depend explicitly on q. Thus, for a large system: $|\sum_q c_q \vec{b}_q e^{-\lambda_q^b \Delta t}| \sim \int_{\lambda_{min}}^{\lambda_{max}} \frac{e^{-\lambda \Delta t}}{\lambda} d\lambda = Ei(\lambda_{max}\Delta t) - Ei(\lambda_{min}\Delta t)$, where $Ei(x)$ is the exponential integral function. For times $\Delta t \to 0$ the integral approaches a constant, which by construction is $|\Delta \vec{n}|$, while for times
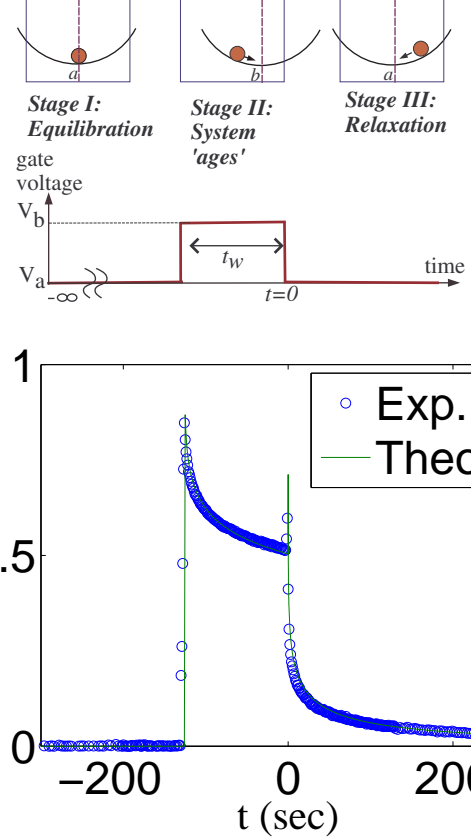

FIG. 1: Schematic demonstration of the aging process. Initially, the system is in a metastable configuration a. When the landscape changes due to a change of gate voltage, the system relaxes towards the new minimum b, during time t_w. When the landscape returns to its initial form, the system relaxes to a. Below we schematically show our results, for the time dependence of the distance to the current metastable state (a or b) $\delta n(t)$. At first, the system sits at a metastable state. At a certain time (-200 sec. for the figure) the system parameter (i.e., gate voltage) is changed, and $\delta n(t)$ jumps and then relaxes logarithmically. At time t_w the system parameter is changed to its initial value, from which the relaxation takes the form $\log(1 + t_w/t)$ (Eq. (2)). The inset shows the experimental result for a typical electron glass aging experiment (courtesy of Z. Ovadyahu). The lower cutoff used in the theory, relevant only for stage II, is a fitting parameter, and was taken to be 50 hours.

$\Delta t \gg 1/\lambda_{min}$ it falls off exponentially. The interesting behavior occurs for the large time window $1/\lambda_{max} < \Delta t < 1/\lambda_{min}$, where we can approximate the integral by $\gamma_E - \log[\Delta t \lambda_{min}]$, with γ_E is the Euler constant. This implies that during stage II we should observe a logarithmic relaxation, which is indeed observed experimentally [13].

In stage III, it is convenient to work in the eigenvector basis of metastable state a. We can express the configuration of the system at the end of stage II as

$\vec{n}(t=0) = \vec{n}_a + \Delta\vec{n} + \sum_q c_q \vec{b}_q e^{-\lambda_q^b t_w}$. Since the perturbation is small, to lowest order we can replace $\vec{b}_q$ by $\vec{a}_q$ and substitute the eigenvector decomposition of Δn, obtaining: $\vec{n}(0) = \vec{n}_a + \sum_q c_q \vec{a}_q (e^{-\lambda_q^b t_w} - 1)$. Note that during the waiting-time, each of the eigenmodes approaches b with a characteristic time $1/\lambda_q$. Therefore only modes obeying $\lambda t_w \gtrsim 1$ had enough time to relax to b. When the voltage is changed to its initial value, this relaxation to b is in fact an *excitation* with respect to the metastable state a. Thus, slowly relaxing modes are also slow to be excited, which is the crux of the matter. The relaxation back to equilibrium during the last stage of the protocol is described as: $\vec{n}(t) = \vec{n}_a + \sum_q c_q \vec{a}_q (e^{-\lambda_q^b t_w} - 1) e^{-\lambda_q^a t}$.

We can approximate $\lambda_q^a \approx \lambda_q^b \equiv \lambda_q$, and obtain for the occupancy vector at time t: $\delta\vec{n}(t) = \sum_q c_q \vec{a}_q (e^{-\lambda_q t_w} - 1) e^{-\lambda_q t}$. Defining $\delta n(t) \equiv |\delta\vec{n}(t)|$, we obtain:

$$\delta n(t) = C[\gamma_E - \log(t+t_w)] - C[\gamma_E - \log(t)] = C \log(1+t_w/t), \quad (2)$$

where $C = \frac{|\Delta\vec{n}|}{\log(\lambda_{max}/\lambda_{min})}$ is a non-universal constant, which depends on the system parameters.

Notice that $|\delta\vec{n}(t)|$ is a function of t/t_w, corresponding to full aging. This is due to the fact that for $f(x) \sim \log(x)$, the difference $f(t + t_w) - f(t)$ is only a function of the ratio t/t_w. It can be proven that *only* a logarithmic function has this characteristic. Since $f(x)$ was essentially a Laplace transform of the eigenvalue distribution, this shows a remarkable relation between the $1/\lambda$ distribution and full aging. Fig. 1 summarizes the theoretically expected time dependence throughout the experiment, and shows the results of a typical electron glass aging experiment.

So far we have discussed the aging of the average occupation numbers. To relate this to the measured observable in the electron glass experiments, the conductance, we need to know the relation $\delta\sigma(\delta n)$. We emphasize that for any such relation we would obtain full aging, since δn has full aging. In the simplest approximation, one would assume a linear relation between the two. In other words $\delta n(t)$, essentially the distance in phase space to the local minimum (the metastable state), is proportional to the excess conductivity. This may arise due to various mechanisms. One may intuitively regard perturbation in occupation number as an effective excess temperature, raising the conductivity. A more subtle mechanism would arise due to slowly relaxing modes, influencing the conductivity of the system via the long-range Coulomb interactions [11, 14]. This is an important point, that may distinguish the electron glass system from spin glasses, and we intend to elaborate on it in future work.

Comparison to experiment.- Fig. 2 compares the theoretical form in stage III and the experimental data of Ovadyahu's group [8]. The experimental data shows a logarithmic regime for times shorter than t_w, and power-

law regime, for times much longer than t_w. These two limits, as well as the crossover between them, are contained in the theoretical expression. The significance of the fit shown in the figure is the linear dependence of the excess conductivity on the predicted function $\log(1+t_w/t)$. There are no fitting parameters other than the proportionality constant, which is non-universal.

The logarithmic behavior predicted for short times is an important distinction between the theory we present and various theories predicting a stretched exponential behavior, $e^{-\left(\frac{t}{t_0}\right)^\alpha}$ [15]. At short times, compared to t_w, the stretched exponential reduces to a power-law, which is inconsistent with the experimental data, as was demonstrated in [13].

For even longer times (on the scale set by t_w), deviations from the theory occur. These can be corrected by taking a non-linear relation between δn and $\delta\sigma$. In order to fit correctly the experimental data, the corrected relation must be sublinear. An additional sublinear term proportional to $\sqrt{\delta n}$ provides excellent correspondence to the experimental data (not shown). That such a sublinear relation may exist is supported by a different experiment, where the conductance is measured as a function of the gate voltage in a relatively fast scan [16, 17].

Generality of the theory.- It is possible that this model is in fact only a caricature of the real scenario: possibly, the system explores many metastable states (local minima of the potential landscape). If we consider the vector of the probabilities to remain in each state, the transition between different minima are also exponentials of a broadly distributed quantity [18]. This implies that the 'ingredients' used in our calculation are still valid: if we consider the vector of probabilities of the system to be in each of the minima, the (classical) dynamics of the system will be described by a master equation of the
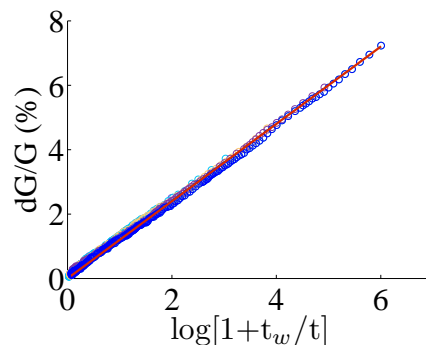

FIG. 2: The excess conductance is plotted as a function of the theoretical expression $\log(1 + t_w/t)$. There are no fitting parameters in this expression. The data shown is for 5 experiments, corresponding to different colors, with different waiting times t_w of 20, 60, 180, 540 and 1620 seconds. The ratio t/tw varies from 0.0025 to 36. We thank Z. Ovadyahu for providing us with the data.

type $\frac{d\vec{P}}{dt} = A\vec{P}$, where $\vec{P}$ is a vector of probabilities to be at different minima. A wide, slowly varying, distribution of barriers would again lead to a $1/\lambda$ distribution of eigenvalues of the matrix A, leading to the relaxation described by Eq. (2).

We demonstrate the generality of the theory by analyzing a different experiment. Ludwig *et al.* [3] measured the dielectric constant of Mylar when pushed out-of-equilibrium, using a similar aging protocol to the one described. We consider the data from the first measurement in each of their aging experiments, for which the system is fully equilibrated. The data presented in the first reference of [3] shows that for times short compared to t_w, the signal is proportional to $\log(ct_w/t)$, with $c = 1.02$ extracted from the data. This fits well with our prediction of $\log(1 + t_w/t)$. We emphasize that in contrast to our theory, the theory used in Ref. [19] gives a non-universal value of c, depending on the system parameters.

Summary.- We have shown how full aging may naturally occur in a model for the electron glass, starting with a microscopic picture, and using a master equation to analyze the dynamics of the system. Under simplifying assumptions, we have shown that the deviation from the metastable state should follow a universal form $\sim \log(1 + t_w/t)$, where t_w is the length of time the perturbation driving the system out-of-equilibrium was on, and t the time of measurement. This agrees well with experimental data of the conductance relaxation in the electron glass InO, within a large time window, ranging from seconds to several hours. The theoretical predictions also explain experiments performed on a totally different glass, the plastic Mylar, where the dielectric constant is measured. Although the system is completely different, the universality of the underlying distribution of relaxation rates $P(\lambda) \sim 1/\lambda$ lead to our general result for the relaxation. This suggests that our model of relaxation arising from a broad $1/\lambda$ spectrum may be a paradigm for aging in various glasses.

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